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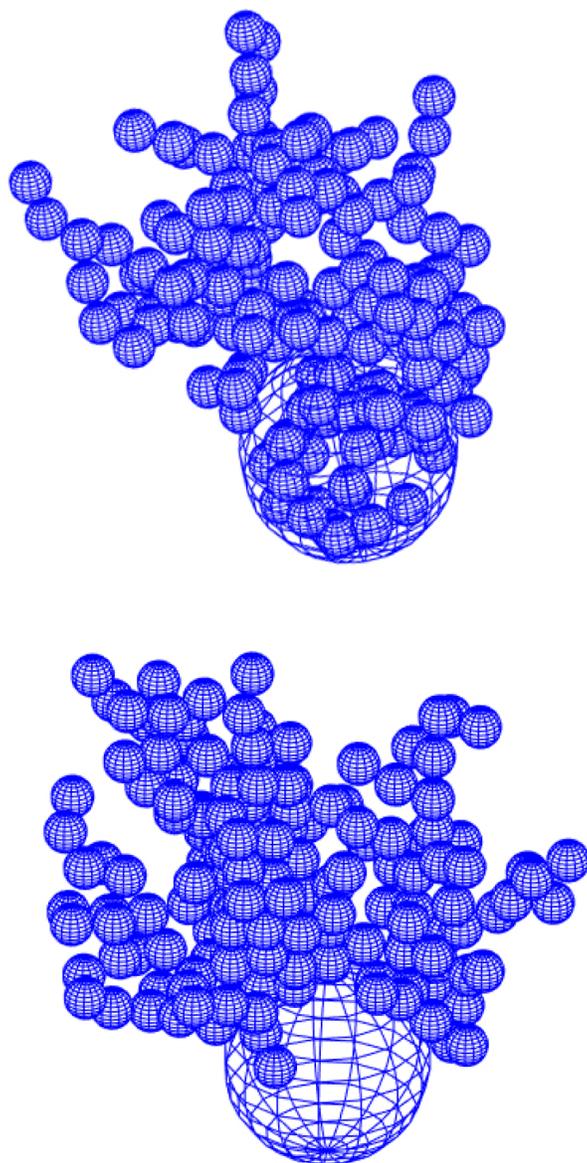
*Supplement of*

## **Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison**

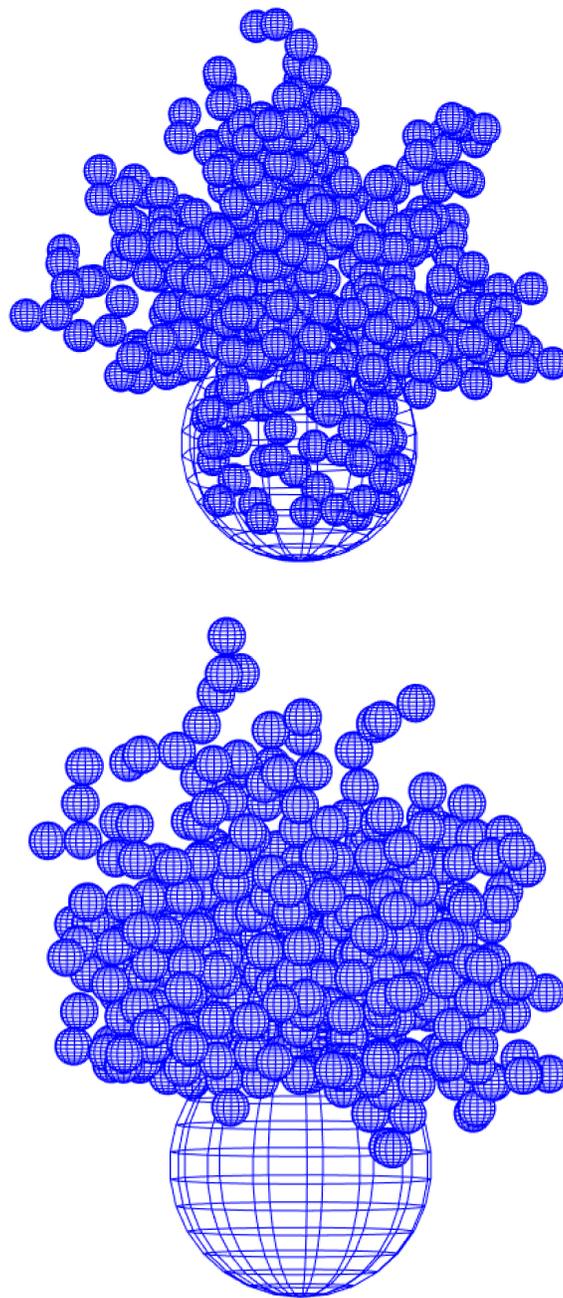
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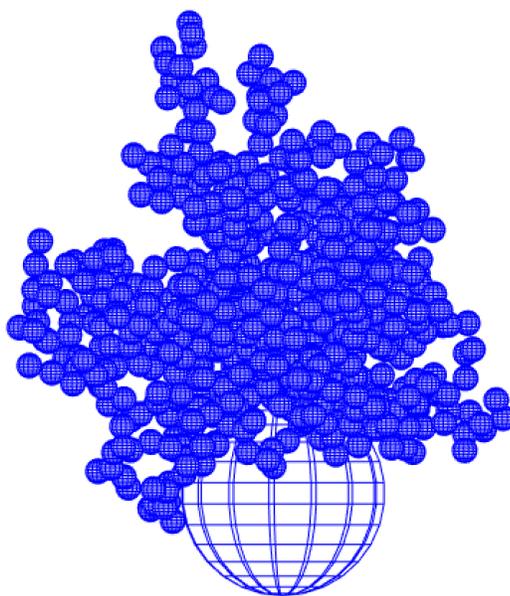
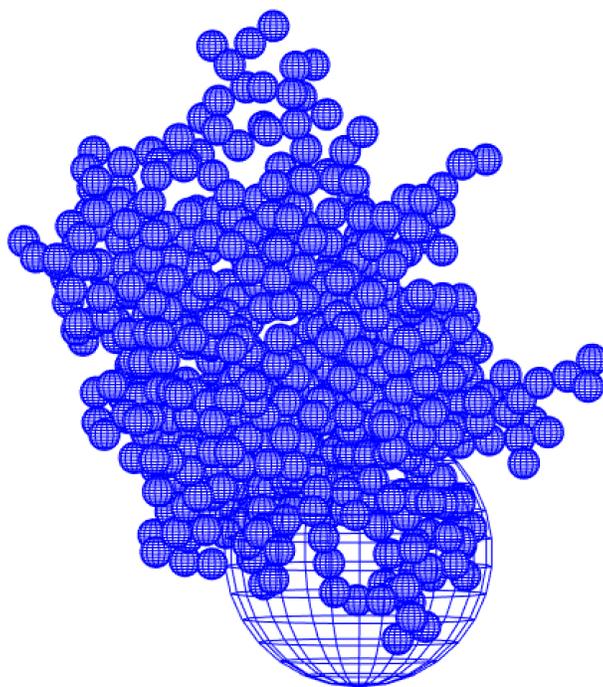
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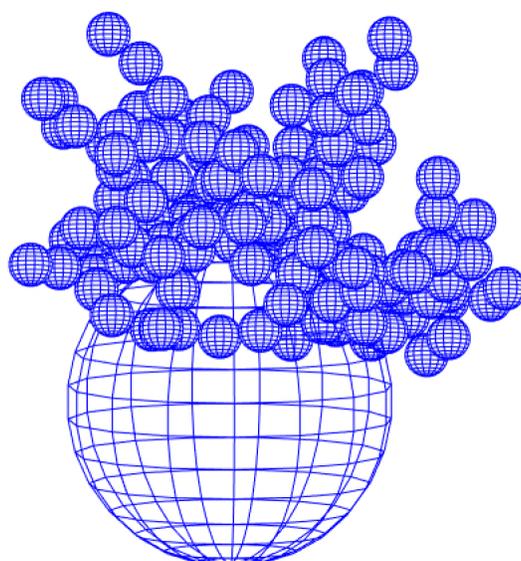
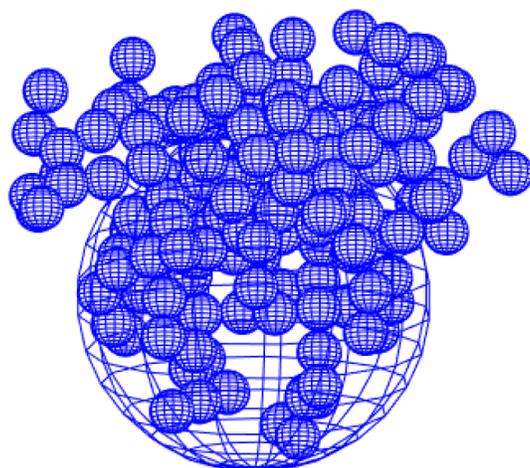
**Figure S1.** The partially encapsulated (top) and externally attached (bottom) coating structures generated by the stochastic procedure used in this study for BC mobility diameter of 155 nm at aging Stage II (i.e., coated by sulfuric acid ( $\text{H}_2\text{SO}_4$ )). The total number of BC primary spherules in each structure is 164, with a diameter of 15 nm for each primary spherule. 30 BC primary spherules are inside the  $\text{H}_2\text{SO}_4$  sphere in the partially encapsulated structure. The diameters of  $\text{H}_2\text{SO}_4$  sphere are 79 nm and 73 nm in the partially encapsulated and externally attached structures, respectively.



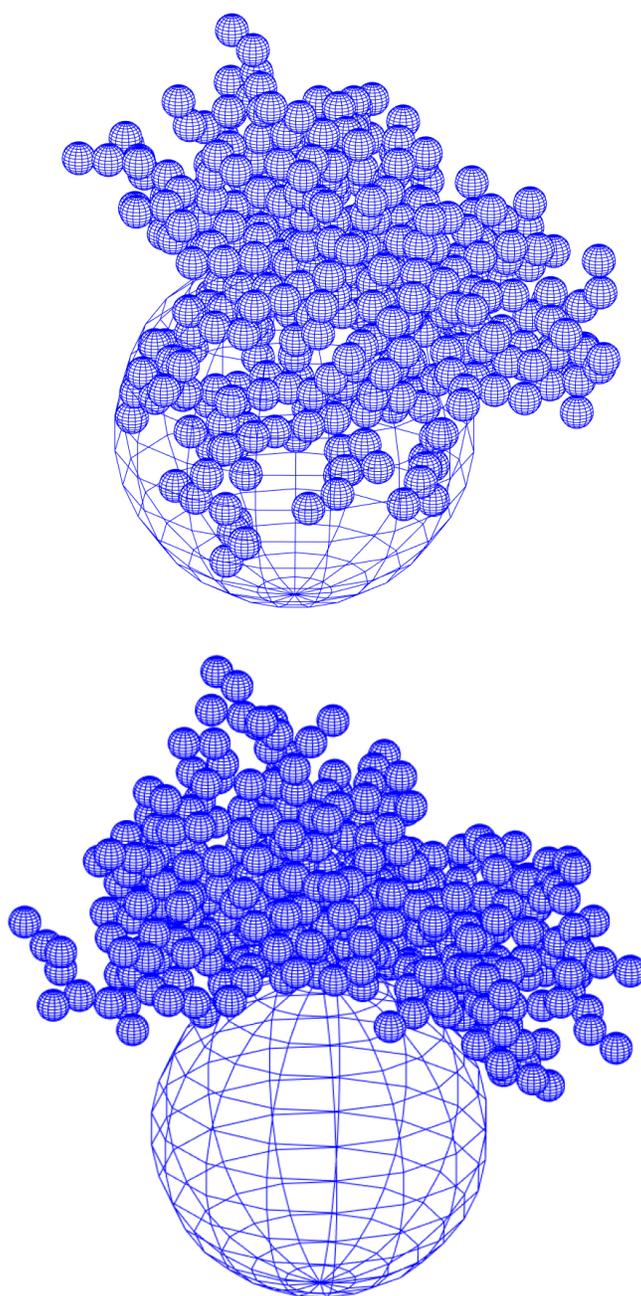
**Figure S2.** Same as Fig. S1, but for BC mobility diameter of 245 nm. The total number of BC primary spherules in each structure is 416. 60 BC primary spherules are inside the H<sub>2</sub>SO<sub>4</sub> sphere in the partially encapsulated structure. The diameters of H<sub>2</sub>SO<sub>4</sub> sphere are 111 nm and 105 nm in the partially encapsulated and externally attached structures, respectively.



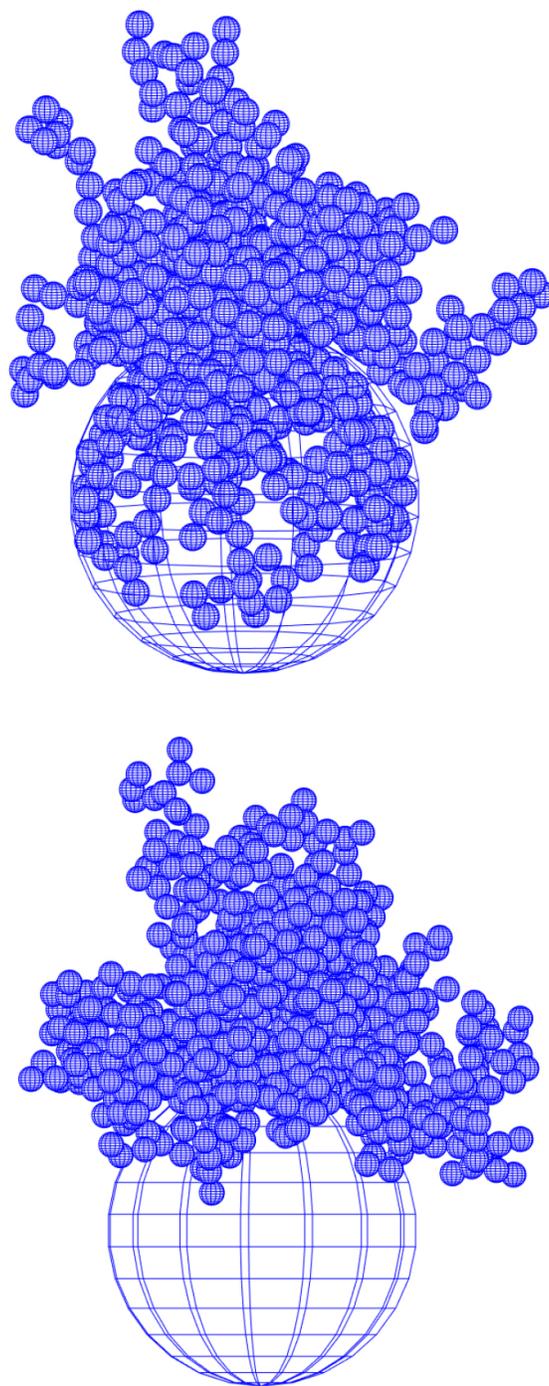
**Figure S3.** Same as Fig. S1, but for BC mobility diameter of 320 nm. The total number of BC primary spherules in each structure is 651. 80 BC primary spherules are inside the H<sub>2</sub>SO<sub>4</sub> sphere in the partially encapsulated structure. The diameters of H<sub>2</sub>SO<sub>4</sub> sphere are 130 nm and 124 nm in the partially encapsulated and externally attached structures, respectively.



**Figure S4.** Same as Fig. S1, but for aging Stage III (i.e., coated by sulfuric acid and water ( $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ )). The total number of BC primary spherules in each structure is 164. 75 BC primary spherules are inside the  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  sphere in the partially encapsulated structure. The diameters of  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  sphere are 119.4 nm and 113.2 nm in the partially encapsulated and externally attached structures, respectively.



**Figure S5.** Same as Fig. S4, but for BC mobility diameter of 245 nm. The total number of BC primary spherules in each structure is 416. 125 BC primary spherules are inside the  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  sphere in the partially encapsulated structure. The diameters of  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  sphere are 163.6 nm and 158.2 nm in the partially encapsulated and externally attached structures, respectively.



**Figure S6.** Same as Fig. S4, but for BC mobility diameter of 320 nm. The total number of BC primary spherules in each structure is 651. 200 BC primary spherules are inside the  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  sphere in the partially encapsulated structure. The diameters of  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  sphere are 192.2 nm and 185.8 nm in the partially encapsulated and externally attached structures, respectively.